Voltage-controlled motional narrowing in a semiconductor quantum dot

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Abstract. We demonstrate the control with a dc-voltage of the environment-induced decoherence in a semiconductor quantum dot embedded in a gated field-effect device. The electrical control of the spectral diffusion dynamics governing the quantum dot decoherence induces various effects, and in particular a narrowing of the quantum dot emission spectrum on increasing the electric field applied to the structure. We develop a model in the framework of the pre-Gaussian noise theory that provides a quantitative interpretation of our data as a function of gate voltage. The standard phenomenology of motional narrowing described in nuclear magnetic resonance is successfully reached by hastening the carrier escape from the traps around the quantum dot through tunnelling under reverse bias voltage. Our study paves the way to a protection of zero-dimensional electronic states from outside coupling through a voltage-controlled motional narrowing effect.

Tunneling is one of the most fundamental manifestations of quantum mechanics. Its applications cover numerous physical situations, and perhaps most importantly in solid state physics for superconductor and semiconductor materials. For instance, electron tunneling between energy bands under a static electric field is the key process for electrical breakdown in bulk materials [1]. In semiconductor superlattices, the reduction of the Brillouin zone along the growth direction allows Zener tunneling between energy minibands at moderate electric fields [2]. In quantum well heterostructures used as optical modulators, field-induced tunneling out of the well controls the population relaxation dynamics and the carrier lifetime decreases on increasing the applied electric field [3, 4].

In this paper, we demonstrate the reduction by tunneling of the environment-induced decoherence in a semiconductor quantum dot (QD) embedded in a gated field-effect device. We show that the QD emission spectrum narrows on increasing the static electric field applied to the structure. This inhibition of the environment-induced dephasing is achieved through tunneling-assisted motional narrowing. Here the motion consists in the tunneling of carriers out of the defects located in the QD mesoscopic environment responsible for spectral noise. We develop a model in the framework of the pre-Gaussian noise theory that provides a quantitative interpretation of our data as a function of temperature and gate voltage.

Our sample consists of a single QD embedded in a field-effect structure, as depicted in Fig. 1(b). Our InGaAs QDs are fabricated by molecular beam epitaxy in the Stransky-Krastanow growth mode. They are separated by a 25 nm GaAs layer from a highly n-doped GaAs substrate. The QDs are capped by 15 nm of GaAs, followed by a 75 nm AlGaAs blocking barrier and finally 60 nm of GaAs. After growth, a 10 nm thick semitransparent Ti Schottky contact (right side of Fig. 1(b)) was deposited on the top surface. In order to study single QDs, we use a 200 nm thick Au mask into which 400 nm diameter apertures are opened lithographically.

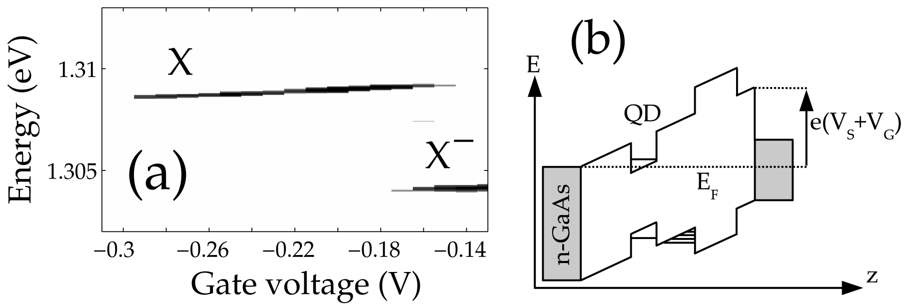

Figure 1. (a) Gray-scale map of the photoluminescence spectrum from a single QD versus gate voltage at 10K. Black (white) corresponds to a signal of 1200 (0) counts per second. (b) Schematic sketch of the heterostructure (under reverse bias) displaying the conduction and valence band edges energy along the growth direction. The applied gate voltage V_G is superimposed on the Schottky voltage V_S of the order of -1.5 V for our structure.

By varying the gate voltage applied between the back and Schottky contacts, we can tune the QD electron states relative to the Fermi energy and thus control the electron occupation in the QDs, as shown in Fig. 1(a). In this figure, we display a gray-scale map of the photoluminescence (PL) spectrum of a single QD as a function of gate voltage, recorded at 10K with a charge coupled device and a spectral resolution of 120 μeV. The PL is non-resonantly excited at 1.96 eV with an excitation density below the QD saturation threshold. For a gate voltage of -0.13 V, the PL spectrum exhibits a resolution-limited line centered at 1.304 eV that is attributed to the radiative recombination of the so called X^- state, formed from two electrons and one hole [5]. By increasing the reverse bias voltage, the PL spectrum is weakly red-shifted because of the quantum confined Stark effect. Around -0.17 V, there is a sudden blue-shift of approximately 5 meV because the PL signal now arises from the neutral exciton, formed from one electron and one hole. This so-called X line is observable for reverse bias voltages up to -0.3 V. For higher electric fields, there is no PL signal anymore because the population relaxation is dominated by the non-radiative processes of electron and hole tunneling out of the QD [5].

The existence of a gate voltage plateau for the neutral exciton line stems from Coulomb blockade since the energies of QD charging by one or two electrons differ by the on-site Coulomb repulsion in the QD [5]. We thus take advantage of the pronounced Coulomb blockade to perform a high resolution study of the X line as a function of the gate voltage. The experimental technique consists of Fourier-transform spectroscopy of the PL signal that allows us to measure both the width and the profile of a single QD emission spectrum, with sub-μeV spectral resolution [6]. In Fig. 2, we display measurements for the neutral exciton line presented in Fig. 1. We show the interferogram contrasts $C(t)$ on semi-logarithmic plots, $C(t)$ corresponding to the modulus of the complex Fourier transform of the signal intensity. In the upper part of Fig. 2, we show our data at 30K for the three gate voltages of -0.24 (a), -0.22 (b) and -0.2 V (c). Our measurements reveal a surprising effect. The coherence decay at -0.24 V (Fig. 2(a)) is slower than at -0.2 V (Fig. 2(c)), and the linewidth decreases with a total reduction of 20% in the whole bias range (Fig. 4(a)). The QD line thus *narrows* on increasing the static electric field in the structure. This original phenomenology is in strong contrast with the well known tunneling-assisted broadening observed in quantum wells, or in QDs in the regime of photo-current spectroscopy [5, 7]. We will see below that the field-induced narrowing observed in Fig. 2 arises from the electrical control of spectral diffusion, and more precisely from motional narrowing assisted by tunneling. In the lower part of Fig. 2, we present the measurements at 10K for the same applied biases. We observe an additional feature that is a strong beating, with a contrast close to unity for -0.22 V (Fig. 2(e)). This means that the PL spectrum consists in that case in a doublet with identical lines. From the beating period, we estimate an energy splitting Δ of the order of 44 μeV. By changing the linear polarization of analysis, there is no modification of the data, thus excluding any effect related to the fine-structure splitting of the neutral exciton line [8]. We will see below that this doublet is again related to spectral diffusion, and that the dependence of the beating contrast on gate voltage is another signature of the electrical control of spectral diffusion.

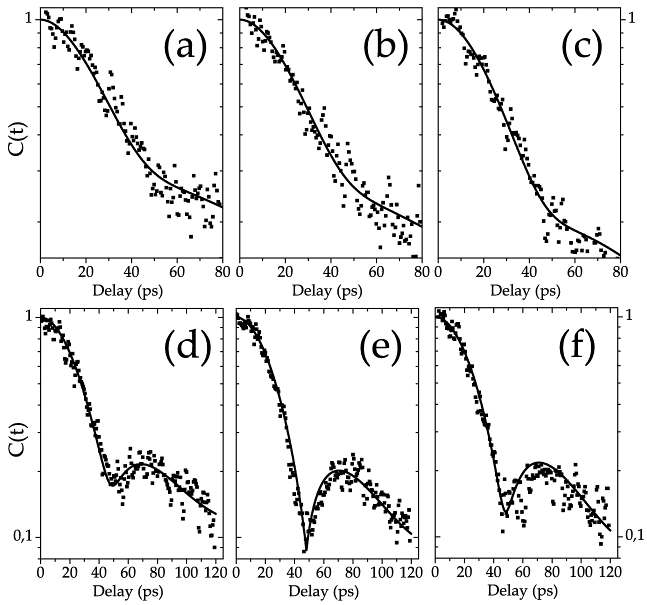

Figure 2. Interferogram contrast $C(t)$ of the PL signal of the single InGaAs QD of Fig. 1, on semi-logarithmic plots, for three gate voltages: -0.24 V at 30K (a) and 10K (d), -0.22 at 30K (b) and 10K (e), and -0.2 V at 30K (c) and 10 K (f). Data (squares), theoretical fits (solid line).

The deviation of the emission spectrum from a lifetime-limited line is to be found in the mesoscopic environment of the QD that generates spectral noise [9, 10, 11]. The presence of localized defects or impurities around the QD influence its emission line through the quantum confined Stark effect: the temporal random fluctuation of the carriers population in these localized traps induce a spectral diffusion of the QD line. In the literature, we can distinguish two extreme cases. The first one is colloidal QDs, or nanocrystals. Spectral diffusion leads in that case to strong perturbation of the spectrum with abrupt fluctuation of the emission energy and also intensity [9]. The second one is encountered for epitaxial QDs, where spectral diffusion may lead to weaker effects. The optical spectrum often consists of a single line, broader than the natural limit of approximately one μeV, but possibly with a Lorentzian profile because of motional narrowing [11]. The original situation depicted in Fig. 2 reveals a rich phenomenology which presents both aspects. In the following, we develop a model in the framework of the pre-Gaussian noise theory that comprises these two aspects of spectral diffusion and allows a quantitative description of our data as a function of gate voltage. Most importantly, we obtain a detailed insight into the tunneling-related processes that modify the QD environment and enable the external control of spectral diffusion with a gate voltage.

The pre-Gaussian noise theory was developed in the context of laser-atom interactions spectroscopy [12]. The spectral noise is represented in the form of a finite number of random-telegraph processes. More precisely, we consider a Markov chain consisting of a superposition of $N+1$ independent two-state jump processes. For an individual jump process, the transition from the upper (lower) state to the lower (upper) one occurs with a probability $dt/T_\downarrow$ ($dt/T_\uparrow$) in the time interval dt and induces a spectral jump -Δ (+Δ). In the context of spectral diffusion in QDs, the upper (lower) state corresponds to an empty (occupied) defect, and $T_\downarrow$ ($T_\uparrow$) to the capture (escape) time of one carrier in the defect. The intensity spectrum related to this elementary Markovian modulation was calculated by Kubo within his stochastic theory of lineshape [13]. From the analytical expression of the optical spectrum [13], we have derived the relaxation function $\phi_1(t)$, which generalizes the treatment of [12, 14] to the case $T_\uparrow \neq T_\downarrow$, and we find:

$$\phi_1(t) = G_{+1}(t) - G_{-1}(t) \tag{1}$$

with

$$G_\varepsilon(t) = \frac{1+\varepsilon Y + i\eta X}{2Y} \exp\left[-\left(1-\varepsilon Y\right)\frac{|t|}{2T_c}\right] \tag{2}$$

where the correlation time T_c is given by $\frac{1}{T_c}=\frac{1}{T_\uparrow}+\frac{1}{T_\downarrow}$, $\eta=\frac{T_\uparrow - T_\downarrow}{T_\uparrow + T_\downarrow}$ characterizes the intensity asymmetry in the emission spectrum, $X=\Delta T_c/\hbar$, and Y is given by $Y^2=1-X^2+2i\eta X$ with the condition $\Re(Y) \geq 0$. If $X \gg 1$ the system is in the slow modulation limit: the optical spectrum exhibits a doublet structure, with a splitting Δ, and the ratio of the integrated intensity of the two separate lines is given by $T_\uparrow/T_\downarrow$. On the contrary, in the fast modulation limit ($X \ll 1$), the doublet structure disappears because motional narrowing induces the spectral coalescence of the two lines [13].

For N independent two-state jump processes, the relaxation function $\phi_N(t)$ is the product of the N single relaxation functions so that $\phi_N(t)=[\phi_1(t)]^N$ [14]. For a large number of two-state jump processes, Wodkiewicz *et al.* showed that the relaxation function $\phi_N(t)$ converges to the one obtained for a Gaussian noise, i.e. for $N \gg 1$, $\phi_N(t)=[\phi_1(t)]^N$ reads [12, 14]:

$$\phi_N(t) \sim \exp\left[-\frac{\Sigma^2\tau_c^2}{\hbar^2}\left(\exp\left(-\frac{|t|}{\tau_c}\right)+\frac{|t|}{\tau_c}-1\right)\right] \tag{3}$$

which is a consequence of the central limit theorem in the context of spectral noise. In Eq.(3), the modulation amplitude Σ corresponds to the standard deviation of the emission energy, that reads $\Sigma=\sqrt{N}\delta\left(\sqrt{\frac{\tau_\uparrow}{\tau_\downarrow}}+\sqrt{\frac{\tau_\downarrow}{\tau_\uparrow}}\right)^{-1}$ [11], where δ is the mean spectral shift of a single jump process, and the average correlation time τ_c is given by $\frac{1}{\tau_c}=\frac{1}{\tau_\uparrow}+\frac{1}{\tau_\downarrow}$. Note that in the following, we will keep the capital letters for the parameters of the relaxation function $\phi_1(t)$. If $\Sigma\tau_c/\hbar \gg 1$, the system is in the slow modulation limit, or stochastic regime [15]: the optical spectrum has a Gaussian profile that reflects the Gaussian distribution law of the spectral noise. If $\Sigma\tau_c/\hbar \ll 1$, motional narrowing leads to a Lorentzian line with a width of $2\Sigma^2\tau_c/\hbar$ [11].

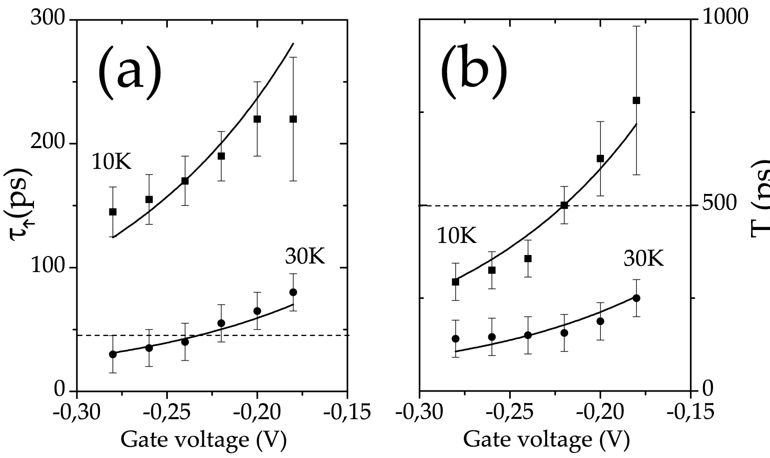

Figure 3. Tunneling times versus gate voltage, for (a) the ensemble of N jump processes, and for (b) the singular one. Data (symbols), theoretical fits (solid line). The horizontal lines indicate the capture times $\tau_\downarrow$= 45 ps in (a) and $T_\downarrow$= 500 ps in (b).

In the framework of the pre-Gaussian noise theory, we take the product $\phi_1(t)\phi_N(t)$, where $\phi_1(t)$ and $\phi_N(t)$ are respectively given by Eq. (1) and Eq. (3), for the relaxation function of our complex mesoscopic environment. This choice means that we separate the N+1 independent two-state jump processes into a large number ($N\gg 1$) of similar ones, and a singular one. The latter is responsible for the strong perturbation leading to the beating and doublet structure, and the former for the broadening of each component. In Fig. 2, we display in solid line the calculated contrasts $C(t)$=$|\phi_1(t)\phi_N(t)|$. We observe an excellent agreement thus showing that our model quantitatively reproduces the whole emission spectrum (shape and width). We fit our complete set of data as a function of voltage and temperature (10K, 30K and 20K not shown here) by taking $\sqrt{N}\delta$=56 μeV, Δ=44 μeV, $\tau_\downarrow$=45 ps, $T_\downarrow$=500 ps, and by only varying $\tau_\uparrow$ and $T_\uparrow$ with the values indicated in Fig. 3(a) and (b), respectively. There is a single exception at 10K and -0.18 V, where we have to take a slightly larger value for $\sqrt{N}\delta$=60 μeV, that we will comment later. We interpret the singular jump process as due to a defect closer to the QD thus resulting in a larger Stark shift ($\Delta\gg\delta$). The constant times $\tau_\downarrow$ and $T_\downarrow$ (shown by dashed lines in Fig. 3) are consistent with multi-phonon assisted capture in defects [16]. The value of 500 ps for $T_\downarrow$ corresponds to its lower limit: for smaller capture times, the beating period would change with gate voltage and temperature, in contradiction with our measurements that show no evidence of motional narrowing for the singular jump process (X=$\Delta T_c/\hbar\gg$ 1), i.e. no coalescence of the doublet structure.

In Fig. 3, we observe a systematic decrease of both escape times $\tau_\uparrow$ and $T_\uparrow$ with the reverse bias, as expected for field-induced tunneling. At 30K, the escape times are smaller than at 10K thus indicating the involvement of phonon-assisted processes since direct tunneling would not be affected by temperature. Note that the consideration of two parallel mechanisms assisted by phonon and tunneling does not allow the temperature activation in Fig. 3 to be reproduced. We thus interpret our data by phonon-assisted tunneling. In analogy to the thermo-ionization of deep centers [17], we take a phonon-assisted tunneling rate that is proportional to the transmission through a triangular barrier due to a static electric field [17]. Namely, we reproduce our data by taking $\tau_\uparrow = \tau_\infty \top(F)^{-1}$ where $\top(F)$ is the barrier transmission given by $\exp\left(\frac{-4\sqrt{2m^* E_i^3}}{3\hbar eF}\right)$ with F the electric field strength along the growth direction, m^* the effective mass and E_i the effective ionization energy. The fitting parameter τ_∞, which corresponds to the mathematical limit of $\tau_\uparrow$ at infinite electric field, is adjusted with temperature. For the singular jump process (Fig. 3(b)), we fit our data with E_i^e=245 meV in the case of electrons (m^*=0.07m_0) or E_i^h=145 meV for holes (m^*=0.34m_0), T_∞=2$\times$10^{-4}ps at 10K and 7.3$\times$10^{-5}ps at 30K. For the large ensemble of N similar jump processes, we reproduce the tunneling time and linewidth variations (Fig. 3(a) and Fig. 4) with E_i^e=235 meV for electrons or E_i^h=138 meV for holes, τ_∞=1.4$\times$10^{-4}ps at 10K and 3.5$\times$10^{-5}ps at 30K. The unfitted data point at 10K and -0.18 V in Fig. 4(b) comes from the fact that we have to take a larger value of $\sqrt{N}\delta$ for this particular case (60 μeV instead of 56 μeV). This additional broadening at the edge of the X plateau is similar to the increased spin dephasing observed in [18]. In our case, the coupling between X and X$^-$ through virtual transitions may increase the Stark shift δ since the negatively charged X$^-$ is much more sensitive to Coulomb effects.

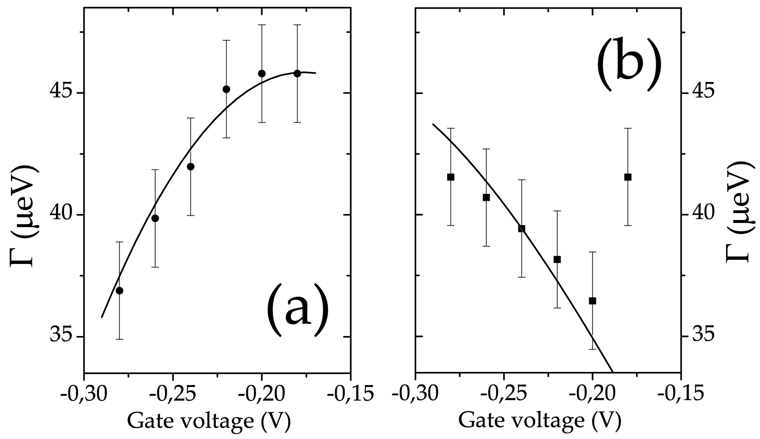

Figure 4. Linewidth versus gate voltage at 30K (a) and 10K (b). Data (symbols), theoretical fits (solid line).

The large values of E_i suggest that the spectral diffusers are deep defects, in agreement with the recent experimental evidence by deep level transient spectroscopy of the coexistence of deep levels with optically active InAs QDs [19]. In Ref. [19] the authors interpret the deep levels as native point defects caused by the strain during the InAs/GaAs growth process, with an areal density comparable to the QD one. In that context, the fast escape may originate from the strong coupling to phonons in point defects [20], and also from a resonant-tunneling effect that can occur for holes because of the presence of two-dimensional holes states in the triangular well at the interface between the capping layer and blocking barrier (Fig. 1(b)) [5]. A complementary structural characterization of these defects is beyond the scope of this work but seems required in order to get a full quantitative analysis of the phonon-assisted tunneling observed in Fig. 3.

We finally focus on the conditions required for achieving field-induced narrowing. We see in Fig. 3(a) that the tunneling time $\tau_\uparrow$ of the large ensemble of N similar jump processes is always larger at 10K than the capture time $\tau_\downarrow$ (shown by a dashed line in Fig. 3(a)), and that the line becomes broader on increasing the electric field (Fig. 4(b)). The interpretation of this effect is to be found in the expression of the modulation amplitude $\Sigma = \sqrt{N}\delta\left(\sqrt{\frac{\tau_\uparrow}{\tau_\downarrow}} + \sqrt{\frac{\tau_\downarrow}{\tau_\uparrow}}\right)^{-1}$, which reaches its maximum value $\sqrt{N}\delta/2$ when $\tau_\uparrow = \tau_\downarrow$, so that a decrease of $\tau_\uparrow$ leads to an increase of the linewidth if $\tau_\uparrow > \tau_\downarrow$. On the contrary, at 30K, $\tau_\uparrow$ crosses the capture time $\tau_\downarrow$, and it turns into a motional narrowing regime where $\tau_\uparrow < \tau_\downarrow$, and the correlation time τ_c decreases while the modulation amplitude is approximately constant. In that case, a decrease of $\tau_\uparrow$ leads to a decrease of the linewidth on increasing the electric field applied to the structure, as shown in Fig. 4(a). This tunnel narrowing effect demonstrates the achievement of the text book phenomenology of nuclear magnetic resonance where the linewidth decreases by increasing the motion. In our solid-state system, the motion is tunneling controlled by a dc-gate voltage, and it induces an inhibition of dephasing in our single QD-device, that could be implemented in electrically-pumped single photon sources [21]. Extrapolation of our data to deeper QDs under higher electric field gives a reduction of spectral diffusion down to the radiative linewidth for a X plateau centered around -0.7 V, that would correspond to an emission energy around 1.2-1.25 eV.

In summary, we demonstrate an inhibition of dephasing by fast tunneling for a single QD device controlled by a gate voltage. The electrical control of the QD mesoscopic environment leads to motional narrowing where the motion consists in carrier tunneling out of the defects around the QD. This work opens the way for a protection of zero-dimensional electronic states from outside coupling with an external bias in field-effect devices.

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